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Date July 31, 2003

J. Lynn Ferry  
J. Lynn Ferry

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

ANDREAS WINTER ET AL.

SERIAL NO: 08/120,105

FILED: SEPTEMBER 10, 1993

FOR: A PROCESS FOR THE PREPARATION OF  
POLYOLEFIN MOLDING COMPOSITIONS  
HAVING A BROAD MELTING RANGE

RECEIVED  
AUG 06 2003  
TC 1700

: ART UNIT: 1713

: EXAMINER: WILSON

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Commissioner for Patents  
Washington, D.C. 20231

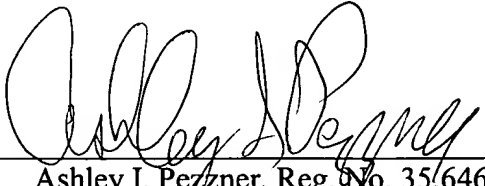
**REPLY TO NON-COMPLIANCE REGARDING APPEAL BRIEF**

In response to the communication mailed July 22, 2003, the applicants have enclosed a new Appendix 1 which also includes claims 22-24. The status of the pending claims was correctly noted in the Appeal Brief which was received in the Patent and Trademark Office on June 25, 2003. However, the PTO was correct that Appendix 1 inadvertently did not include claims 22-24. The applicants have enclosed the correct claims in the accompanying Appendix 1. The applicants respectfully request that the enclosed Appendix 1 be substituted for the previously submitted Appendix 1.

It is believed that no fees are due, however, in the event that the undersigned is mistaken in his calculations, an appropriate extension of time to respond is respectfully petitioned for, and the Commissioner is hereby authorized to charge the account of the undersigned attorneys, Patent Office Deposit Account No. 03-2775, for any fees which may be due upon the filing of this paper.

Respectfully submitted,

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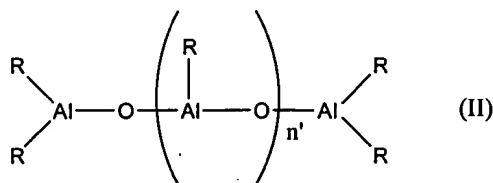
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APPENDIX 1RECEIVED  
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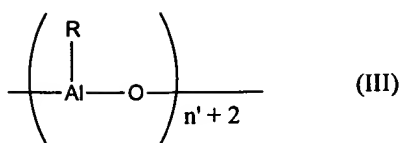
15. The process as claimed in claim 17, wherein the metallocenes are selected from the group consisting of  $\text{rac-Me}_2\text{Si(2-methyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-Me}_2\text{Si(indenyl)}_2\text{HfCl}_2$ ,  $\text{pheny(methyl)Si(2-methyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{Me}_2\text{Si(2-methyl-4-phenyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{Me}_2\text{Si(2-methyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{Me}_2\text{Si(indenyl)}_2\text{HfCl}_2$ ,  $\text{phenyl(methyl)silyl(indenyl)}_2\text{HfCl}_2$ ,  $\text{rac-ethylene(2-methyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-Me}_2\text{Si(2-methyl-4-phenyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-ethylidene(2-methyl-4,6-diisopropyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-Me}_2\text{Si(2-methyl-4,5-benzoindenyl)}_2\text{ZrCl}_2$ ,  $\text{dimethylmethylene (9-fluorenyl)(cyclopentadienyl)ZrCl}_2$ ,  $\text{phenyl(methyl)methylene(9-fluorenyl)(cyclopentadienyl)ZrCl}_2$ ,  $\text{rac-phenyl(methyl)silyl(2-methyl-4,6-diisopropyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{Ph(Me)Si(2-methyl-4-phenyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-Me}_2\text{Si(2-methyl-4-(1-naphthyl)-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-Me}_2\text{Si(2,5,6-trimethyl-1-indenyl)}_2\text{ZrCl}_2$ ,  $\text{rac-Me}_2\text{Si(4,5-benzo-1-indenyl)}_2\text{ZrCl}_2$  and  $\text{rac-Me}_2\text{Si(4-phenyl-1-indenyl)}_2\text{ZrCl}_2$ .
17. A process for the preparation of a polyolefin molding composition comprising at least two polyolefinic components, wherein the composition is characterized by a broad, bimodal, or multimodal melting range in a DSC spectrum determined with

a heating/cooling rate of 20° C/min wherein the peak in the melting range has a maximum and can be bimodal or multimodal and the maximum of the peak in the melting range is between 120 and 165°C, the half-intensity width of the melting peak is broader than 10°C and the width determined at quarter peak height is greater than 15°C, wherein such process comprises the direct polymerization of propylene or copolymerization of propylene with olefins of the formula  $R^aCH = CHR^b$ , in which  $R^a$  and  $R^b$  are identical or different and are a hydrogen atom or an alkyl radical having 2 to 14 carbon atoms wherein the polymerized ethylene content of the resulting polyolefin composition is from 0 to 2.5% by weight, to at least two polyolefins of different melting points, wherein the melting points of the polyolefins must differ by at least 5° C, and wherein the polymerization is carried out at a temperature of from -60 to 200°C, and a pressure of from 0.5 to 100 bar, in solution, in suspension or in the gas phase, in the presence of a catalyst, wherein the catalyst comprises

(A) at least two racemic or s-symmetric metallocenes as transition-metal components and an aluminoxane of the formula II



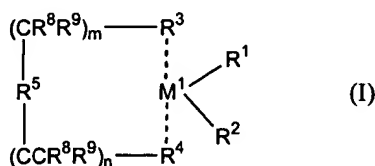
and/or of the formula III



where in the formulae II and III, the radicals R may be identical or different are a C<sub>1</sub>-C<sub>6</sub>-alkyl group, a C<sub>1</sub>-C<sub>6</sub>-fluoroalkyl group, a C<sub>6</sub>-C<sub>18</sub>-aryl group, a C<sub>6</sub>-C<sub>18</sub>-fluoroaryl group or hydrogen, and n' is an integer from 0 to 50, and the aluminoxane component may additionally contain a compound of the formula AlR<sub>3</sub>, or

(B) at least two racemic or s-symmetric metallocenes as transition-metal components and a salt-like compound of the formula R<sub>x</sub>NH<sub>4</sub> · x or of the formula R<sub>3</sub>PHBR'<sub>4</sub> wherein x is 1, 2 or 3, R is identical or different and is alkyl or aryl, and R' is aryl, which may also be fluorinated or partly fluorinated,

where the transition-metal component used comprises at least two metallocenes of the formula I:



in which

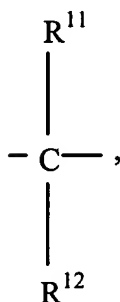
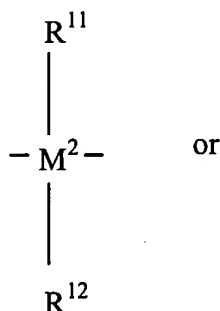
M<sup>1</sup> is Zr or Hf,

R<sup>1</sup> and R<sup>2</sup> are identical or different and are a hydrogen atom, a C<sub>1</sub>-C<sub>10</sub>-alkyl group, a C<sub>1</sub>-C<sub>10</sub>-alkoxy group, a C<sub>6</sub>-C<sub>10</sub>-aryl group, a C<sub>6</sub>-C<sub>10</sub>-aryloxy

group, a C<sub>2</sub>-C<sub>10</sub>-alkenyl group, a C<sub>7</sub>-C<sub>40</sub>-arylalkyl group, a C<sub>7</sub>-C<sub>40</sub>-  
alkylaryl group, a C<sub>8</sub>-C<sub>40</sub>-arylalkenyl group, or a halogen atom,

R<sup>3</sup> and R<sup>4</sup> are identical or different and are indenyl, cyclopentadienyl or fluorenyl  
which are optionally substituted with substituents as defined for R<sup>11</sup> and  
R<sup>12</sup> and where the substituents are identical or different or form together  
with the atoms connecting them a ring,

R<sup>5</sup> is



where R<sup>11</sup> and R<sup>12</sup> are identical or different and are a hydrogen atom, a halogen  
atom, a C<sub>1</sub>-C<sub>10</sub>-alkyl group, a C<sub>1</sub>-C<sub>10</sub>-fluoroalkyl group, a C<sub>6</sub>-C<sub>10</sub>-aryl  
group, a C<sub>6</sub>-C<sub>10</sub>-fluoraryl group, a C<sub>1</sub>-C<sub>10</sub>-alkoxy group, a C<sub>2</sub>-C<sub>10</sub>-alkenyl  
group, a C<sub>7</sub>-C<sub>40</sub>-arylalkyl group, a C<sub>8</sub>-C<sub>40</sub>-arylalkenyl group or a C<sub>7</sub>-C<sub>40</sub>-

alkylaryl group, or  $R^{11}$  and  $R^{12}$  together with the atoms connecting them,  
form a ring,

$M^2$  is silicon or germanium,

$R^8$  and  $R^9$  are identical or different and are as defined for  $R^{11}$  and

m and n are identical or different and are zero or 1 and wherein for at least one of  
the at least two metallocenes  $R^3$  is a substituted indenyl or an optionally  
substituted fluorenyl.

18. The process as claimed in claim 17, wherein the process comprises the direct polymerization of propylene or copolymerization of propylene with an olefin selected from the group consisting of ethylene, 1-butylene, 1-hexene, 4-methyl-1-pentene, 1-octene and mixtures thereof.
19. The process as claimed in claim 17, wherein  $R^1$  and  $R^2$  are identical or different and are a  $C_1$ - $C_{10}$ -alkyl group, a  $C_1$ - $C_{10}$ -alkoxy group, a  $C_6$ - $C_{10}$ -aryl group, a  $C_6$ - $C_{10}$ -aryloxy group or halogen.
21. The process as claimed in claim 20, wherein said two different metallocenes are rac-dimethylsilyl(2-methyl-1-indenyl)<sub>2</sub>ZrCl<sub>2</sub> and rac-dimethylsilyl(indenyl)<sub>2</sub>HfCl<sub>2</sub>.
22. The process as claimed in claim 17, wherein said two different metallocenes are rac-phenylmethylsilyl(2-methyl-1-indenyl)<sub>2</sub>ZrCl<sub>2</sub> and rac-dimethylsilyl(2-methyl-4-phenyl-1-indenyl)<sub>2</sub> ZrCl<sub>2</sub>.
23. The process as claimed in claim 20, wherein said two different metallocenes are rac-phenylmethylsilyl(indenyl)<sub>2</sub>HfCl<sub>2</sub> and rac-dimethylsilyl(2-methyl-4-phenyl-1-indenyl)<sub>2</sub> Zr Cl<sub>2</sub>.

24. The process as claimed in claim 17, wherein  $R^1$  and  $R^2$  are identical or different and are a hydrogen atom, a  $C_1$ - $C_3$ - alkyl group, a  $C_1$ - $C_3$ -alkoxy group, a  $C_6$ - $C_8$ -aryl group, a  $C_6$ - $C_8$ -aryloxy group, a  $C_2$ - $C_4$ -alkenyl group, a  $C_7$ - $C_{10}$ -arylalkyl group, a  $C_7$ - $C_{12}$ -alkylaryl group, a  $C_8$ - $C_{12}$ -arylalkenyl group, or chlorine  
 $R^{11}$ ,  $R^{12}$  and  $R^{13}$  are identical or different and are a hydrogen atom, a  $C_1$ - $C_4$ - alkyl group,  $CF_3$  group, a  $C_1$ - $C_4$ -alkoxy group, a  $C_6$ - $C_8$ -aryl group, pentafluorophenyl group, a  $C_2$ - $C_4$ -alkenyl group, a  $C_7$ - $C_{10}$ -arylalkyl group, a  $C_7$ - $C_{12}$ -alkylaryl group or a  $C_8$ - $C_{12}$ -arylalkenyl.
25. The process as claimed in claim 17, wherein  $R^1$  and  $R^2$  are identical and are methyl or chlorine,  
 $R^4$  and  $R^3$  are indenyl, cyclopentadienyl or fluorenyl, where these ligands may carry additional substituents as defined for  $R^{11}$ .
27. The process as claimed in claim 17, wherein said metallocenes are chiral metallocenes.
28. The process as claimed in claim 25, wherein  $-(CR^8R^9)_m-R^5-(CR^8R^9)_n$  is ethylene or  $CH_3SiCH_3$ .
29. The process as claimed in claim 17, wherein  $M^1$  is Zr for all the metallocenes of formula 1.
30. The process as claimed in claim 17, wherein the polyolefin molding composition is a homo or copolymer with a propylene content of from 97.5 to 100% by weight.



31. The process as claimed in claim 17, wherein the polyolefin molding composition is a homo polymer of propylene or a propylene ethylene copolymer with an ethylene content of up to 2.5% by weight.
32. The process as claimed in claim 17, wherein the composition is characterized by a broad melting range.

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